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Intense Plasma-Waveguide Terahertz Sources for High-Field THz probe science with ultrafast lasers for Solid State Physics,

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INTENSE PLASMA-WAVEGUIDE TERAHERTZ SOURCES

PI: S R Andrews
November 2014-November 2015

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Summary
This report describes progress in exploiting a new experimental capability enabled by the award, namely the ability to excite material with intense and tunable femtosecond pulses of near infrared radiation using an optical parametric amplifier (OPA). This tool, which was installed in February 2015 as an add-on to an existing high energy laser system, has been applied to the study of intense terahertz radiation generated in gaseous plasmas in purpose fabricated hollow core fibres. Using the OPA we have also studied high intensity THz generation in an organic crystal and optical nonlinearities in graphene and have made a quantitative comparison different terahertz detection modalities for nonlinear terahertz spectrocopy.

1. Introduction

The initial aim of this 12 month project was to investigate terahertz (THz) generation in laser ionized gases confined in hollow silica waveguides, exploiting the ability of the optical parametric amplifier (OPA) purchased with the aid of the grant to provide tunable femtosecond excitation pulses in the 1.2-1.8 μm range when pumped by our existing 2.5 mJ, 800 nm regenerative amplifier. This was a small award for equipment only and short proof of principle investigations. The main rationale for the project is that there is a growing need for more intense and efficient pulsed THz sources for nonlinear spectroscopy, stand-off chemical sensing and video rate THz imaging. The particular waveguides of interest are similar to microstructured optical fibres (MOFs) recently developed in Bath for use at wavelengths of a few microns but made on a larger scale because of the much longer wavelength at THz frequencies. The project has progressed more slowly than hoped, partly because of a manpower shortage and partly because the computer model of 2-colour THz generation and propagation in waveguides that we have developed to evaluate fibre designs before fabrication is only one dimensional and this now appears too simplistic. However, we have been able to create plasmas inside hollow waveguides and make changes to the THz spectrum by adjustment of the phase between the fundamental and second harmonic pump pulses as a result of the waveguide dispersion. We have also taken advantage of the longer laser wavelengths now available to explore in parallel the generation of intense THz radiation in an organic crystal with very high second order nonlinearity and second and third order optical nonlinearities in optically pumped graphene and thin metal films. We have also made a comparison of different coherent detection techniques that can be used with high intensity THz sources. In the following sections we describe these developments in or detail.

2. Two colour excitation of intense THz radiation in MOFs

The generation of THz radiation in 2-colour (first and second harmonic) laser ionized air was first reported by Cook and Hochstrasser [1]. Because there is no significant THz absorption in dry air, the THz spectrum is relatively featureless compared with difference frequency generation in a nonlinear crystal and there is no damage threshold to limit up-scaling of the THz power. However, the optical to THz conversion efficiency is < 0.01% when pumping at 800 nm and 400 nm. Since then several groups have incrementally improved the efficiency by optimising the phase matching and group delay between the pump first and second harmonics but this has yielded only a factor of two improvement. The energy conversion efficiency increases as approximately the 4^{th} or 5^{th} power of the pump wavelength [2] so that the longer wavelengths produced by the OPA offer opportunities. In our experimental setup we typically generate 180 nJ THz pulses with 15 THz bandwidth starting with 2 mJ, 800 nm pump pulses and using air as the nonlinear medium. With 1300 nm and 650 nm pumping we generate 40

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nJ THz pulses with similar bandwidth using a 0.28 mJ primary pump energy. Several characteristics limit the usefulness of this source for nonlinear THz spectroscopy:

i) radiation is generated in a cm or more long filament arising from the interplay of self–focusing and defocusing due to plasma formation. Such a 'line' source is difficult to focus to the desired wavelength cubed volume in order to obtain the desired very strong THz fields (≥ 1 MV/cm).
ii) the spectrum is broad so that the spectral intensity at a particular frequency is generally smaller than desired. Some spectral tunability would be useful.
iii) the interaction length determining the optical down conversion efficiency is limited by the coherence length which is of only of order 1 cm in air.

The project had the aim of exploring the generation of THz radiation in an air filled MOF in which the phase matching and spectrum could be modified by design so as to achieve a longer interaction length and thus higher conversion efficiency, spectral tunability and THz source with the characteristics of a Gaussian waveguide mode that could be imaged on to a sample or detector more efficiently. The OPA offered several advantages for this work: the long wavelength gives significantly higher (x7 or 8) conversion efficiency so that less energetic laser pulses can be used with lower associated damage to the waveguide walls and the ability to tune the excitation wavelength allows some tuning of phase matching via the modal dispersion in the waveguide. The type of MOF envisaged is a coaxial structure comprising a near infrared/visible fibre with an air core of order 200 µm diameter suspended inside a larger (few mm diameter core) THz guide. An optical micrograph showing a cross section of a typical fused silica MOF that we have explored is shown in Fig. 1. Unlike the bandgap guiding of conventional hollow core photonic crystal fibre, the mechanism here anti-resonant reflective guiding with inhibited coupling between the core and cladding modes due to a high degree of transverse field mismatch arising from localisation of the cladding mode fields in the fine silica struts. We have manufactured such fibres using stack and draw techniques in the fibre fabrication facility in Bath. Fibres can be drawn to have a small core diameter for use as a near infrared/visible central guide where the THz radiation is generated or a large diameter for a coaxial THz guide. Important features are the thin outer jacket and the very thin walls. Both of these are requirements for low overlap of guided waves with silica and thus broadband guiding with low dispersion and loss. The THz fibres are surprisingly robust and retain some tolerance to bending despite the large dimensions. THz pulse propagation can be characterised by THz waveguide imaging using a scanning photoconductive near field probe [3] at low frequency, as illustrated in Fig. 1.

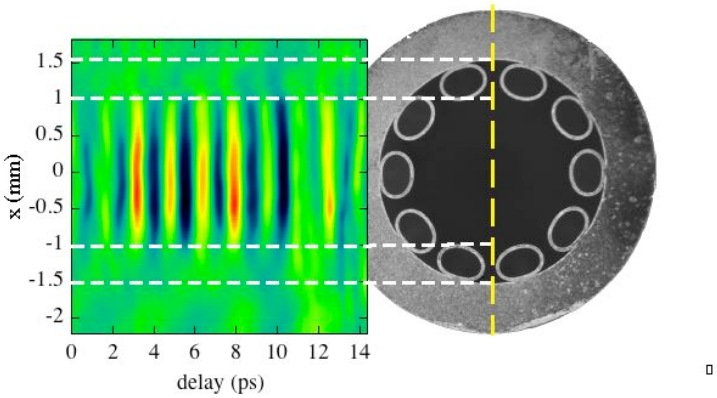

Fig. 1: Space time THz waveguide image of pulse propagation in the MOF with optical micrograph cross section shown.It is clear that radiation is largely confined to the region inside the ring of thin wall silica tubes.

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Using the 'air-biased coherent detection' technique, which is actually based on four wave mixing in butane in our case, THz generation and guiding can be studies at frequencies beween ~ 1 and 30 THz. Fig. 2 shows spectra obtained from a plasma excited inside a 1.5 mm hollow core fibre with average wall thickness of 15 μm. The core is sufficiently large that there is little waveguiding of the pump light but the THz light is guided well over short distances of order 10 cm and a bandwidth of ~ 20 THz. Guiding breaks down when the wavelength becomes larger than about $1/5^{th}$ of the core diameter i.e. when the frequency is less than ~ 1 THz. There is also resonant loss at frequencies where there is resonant Fabry-Perot-like transmission through the thin silica tubes surrounding the core. The thinnest silica struts are about 10 μm in thickness. The spectrum can be altered by changing the relative phase of the two pump pulses. This is presently being investigated further to see if it is possible to engineer larger, controlled changes.

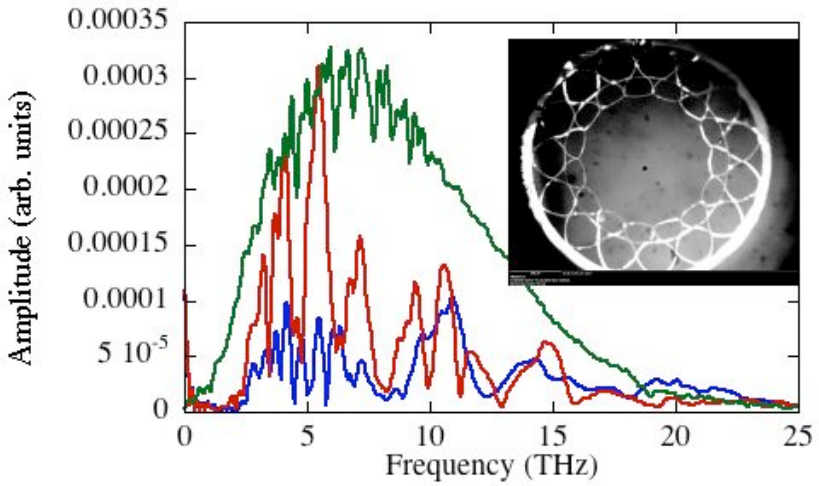

Fig. 2: Effect of relative phase of pump pulses on THz spectrum generated by 2-colour excitation of an air plasma in the 20 cm long silica waveguide shown in the inset. The red and blue spectra are recorded with a π/2 relative phase shift between the fundamental and second harmonic pump pulses. The green curve shows the spectrum if the waveguide is removed (not on same scale). The fine structure evident in the green curve is an artefact due to reflections in a thin diamond window on the detector.

3. Optical nonlinearities in graphene

The very high 3rd order nonlinearity in graphene was first observed using mid infrared laser pulses but the estimated magnitude of the nonlinear optical coefficient is at odds with theoretical predictions. We have used 1300 nm radiation from the OPA and detected THz emission from 2 colour excitation which creates interfering photocurrents. Using the fact that the THz emission amplitude is proportional to the product of sample thickness 3rd order nolinear optical coefficient and by comparing with the magnitude of emission from a thin gold film, we find a value of $X^{(3)} \sim 2{,}0 \pm 0.5 \times 10^{-7}$ esu, which is 50 times larger than for gold and close to that found in ref 4. using very different wavelengths. Our experiments, in which we have compared CVD graphene monolayers obtained from 3 different sources with a variety of gold films, therefore confirm the previous measurement.

We have also studied THz emission associated with the photon drag effect in single layer CVD graphene pumped at 1300 nm and find similarities with the optically excited emission from thin metal films under similar excitation conditions. This leads us to the conclusion that the photon drag effect is also important in understanding THz emission from metal films, a fact not previously appreciated. Although our results on graphene are similar to previous

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measurements at much lower pulse energy [5], there are some important differences which could be associated with the much higher photoexcited carrier density in our measurements. These differences are the subject of ongoing investigations,

4. Intense terahertz generation on OH-1

The non-ionic organic crystal known as OH-1 (2-[3-(4-hydroxystyryl)-5.5-dimethylcyclohex-2-enylidene] malonitrile)) has been reported to be a highly efficient source of few THz radiation when excited under quasi phase matched conditions near 1300 nm with mJ pulses due to its very high second order nonlinear coefficient d_{111} and low THz absorption between 0.3 and 3 THz [6]. Conversion efficiencies exceeding 1% have been reported by a Swiss group at the Paul Scherrer Institute working with the crystal growers but this has not been confirmed independently. These results are interesting because the collinear experimental geometry is very simple and currently the only other benchtop source for nonlinear THz spectroscopy at around 1 THz is Mg doped $LiNbO_3$ excited in a complicated tilted-wavefront geometry. The conversion efficiency in the latter case is typically of order 0.1% and the frequency is limited by phonon absorption to below 1 THz. We have obtained an OH-1 crystal from the Swiss growers and reproduced the reported experimental conditions and indeed find a very high conversion efficiency of order 1 or 2 %. However, only 10% of the radiation generated seems to be in the few THz range, which seems to contradict the published results. We have used thermopiles independently calibrated by two different methods to measure the THz pulse energy. It is presently unclear where the remaining radiation is spectrally located or what its origin is but is blocked by a 20 THz low pass filter. This additional component is strongly polarised in the same way as the THz radiation and has a power dependence consistent with a second order nonlinear process so that its origin cannot be simply thermal. Two points that might be important are that the crystal absorption is strongly polarization dependent and a maximum when the THz conversion efficiency is also a maximum and our laser pulse repetition rate is 20 times higher than used by the Swiss group. It is possible therefore that there is some significant change in crystal properties with temperature. Indeed, the thermal relaxation time is estimated to be as long as 10 ms. The most likely problem is a change in the refractive index, which will affect the efficiency of phase matching. This will be the subject of further investigation. At present we can obtain 200 nJ pulses near 2 THz with the OH-1 crystal when exciting with 300 μJ, 1300 nm, 65 fs pulses. When tightly focused, this allows the creation of peak THz electric fields of ~0.5 MV/cm. Although good, we believe that cooling the crystal could lead to a 3 or 4-fold improvement. The THz spectra produced by OH-1 and a laser ionized plasma co-excited by 1300 nm and 650 nm pulses are shown for comparison in Figs. 3 and 4.

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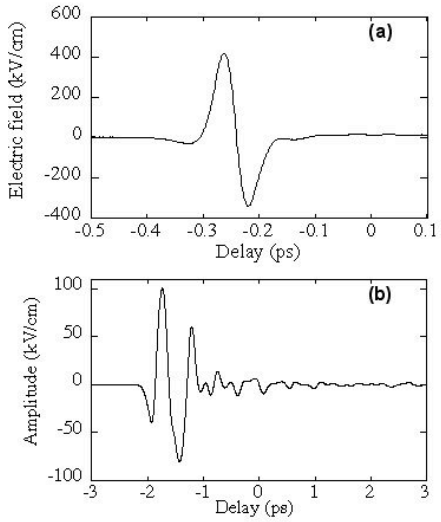

Fig. 3: Time domain traces of (a) a 2-colour excited air plasma and (b) a 450 µm thick OH-1 crystal excited at 9 mJ/cm2. (a) was obtained by air biased coherent detection and (b) by electro-optic sampling in a 40 µm thick ZnTe crystal. Fundamental excitation wavelength is 1300 nm in both cases. Detection wavelength is 800 nm.

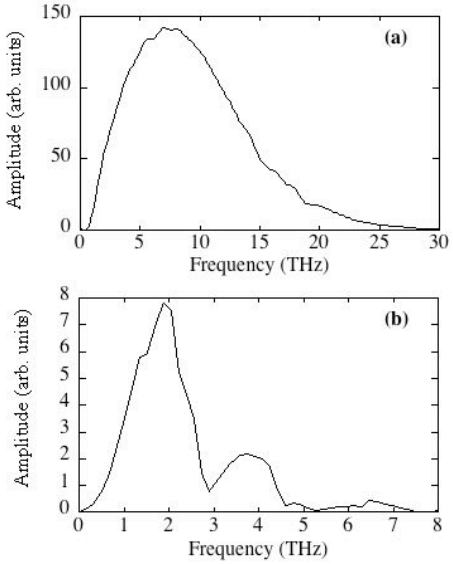

Fig. 4: THz emission spectra of (a) a 2-colour excited air plasma and (b) a 450 µm thick OH-1 crystal excited at 9 mJ/cm2. THz pulse energy in (a) is 50 nJ after a Ge filter. In (b) it is 200 nJ after a PTFE filter. Pump energy is 300 µJ.

5. Comparison of coherent detection techniques for use in nonlinear THz spectroscopy

The frequency response of ion implanted silicon photoconductive devices designed for coherent detection in time domain terahertz spectroscopy has been studied between 0.2 and 30 THz. Unlike devices using polar photoconductors or ones having polar substrates, which have a complicated response spectrum in the region of their restrahlen bands, the response of silicon detectors fabricated on silicon substrates is relatively featureless. When used with amplified laser systems the dynamic range of Si detectors is shown to be very similar to that of GaAs devices with the same geometry over a 20 THz range, superior to air biased coherent detection (ABCD) at frequencies below ~7 THz and comparable with both ABCD and

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electro-optic sampling in thin ZnTe crystals between 7 and 20 THz. Together with their ease of use and linear response to terahertz fields approaching 1 MV/cm, this suggests that Si photoconductors could be a competitive choice for sensitive detection in nonlinear hyper-terahertz spectroscopy. Supporting data is shown in Fig. 5. Results of this work have been published in ref. 7.

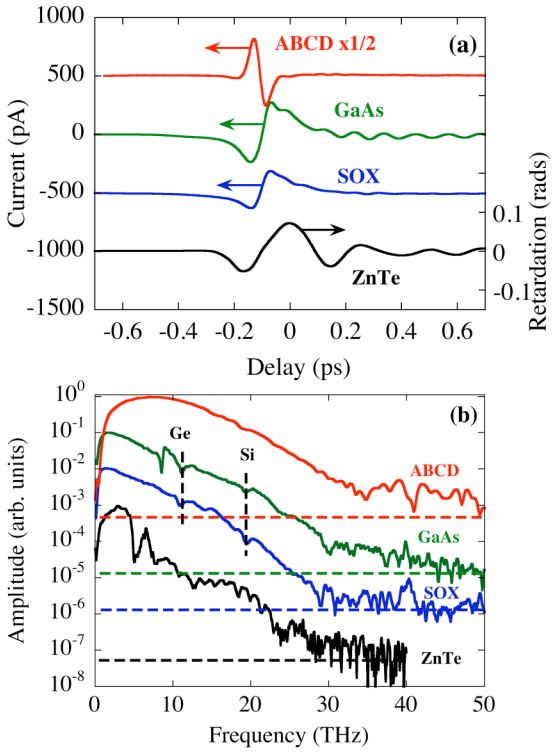

Fig. 5: Time domain detector signals obtained from a 2-colour laser ionized plasma source using different detectors. (b) The corresponding spectra and average background levels in the absence of a THz signal (horizontal dashed lines). The spectra have been normalised to different powers of ten for clarity. The vertical dashed lines in (b) indicate frequencies of multiphonon absorption in Ge and Si filters.

5. Conclusions

The OPA has proved itself a useful tool in the evaluation and development of high intensity THz sources and associated detectors and in probing fundamental THz properties of materials. The research described in sections 2 to 4 is incomplete but we expect that it will result in publications over the next 12 months, which shall of course properly acknowledge EOARD support.

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